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ELASTIC COEFFICIENTS AND INTERNAL FRICTION OF SILICON MONOCRYSTAL SPHERES

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ABSTRACT

Using mechanical-spectroscopy methods, we measured the elastic-stiffness coefficients of four spheres of high-quality monocrystal silicon. Within 1 part in 1000, the C_{ij} agreed with that reported by Bell Laboratories more than thirty years ago. We attribute the internal friction Q_{ij}^{-1} mainly to dislocations. Departure from the Koehler-Granato-Lucke vibrating-string model suggest dislocation-kink motion.

TRANSCRIPT

DR. LEDBETTER: Good morning.

[Transparency 1]

This morning I shall be talking about this topic and I shall focus mainly on the internal friction part, because it is no surprise that the elastic constants of silicon have been studied since the early 1960s and are quite well understood.

[Transparency 2]

The silicon that we studied is a very special silicon. It is part of an international program involving Italy, the United States, Germany, Japan, and Australia. All of these people say it is the best silicon ever made in terms of impurity content and dislocations.

The goal of that program is to propose a new mass standard, an international mass standard of silicon. It also involves some very interesting basic physics about what is the molecular weight of silicon and how the kilogram standard and Avogadro's number come into this. Actually, the existing uncertainties in Avogadro's number are a big problem for these people.

[Transparency 3]

Our specimens are shown here. We studied a total of 5 spheres. These lower spheres are 1 cm in diameter. Two of them are fine ground, one of them is polished. I mention that, because

we found no difference in their elastic constants (these were all crystals from the same boule of silicon and these happened to come from Germany).

These upper 2 spheres came from Japan and, of course, the Japanese think their silicon is best and the Germans think their silicon is best. We also complemented these measurements with some rod-resonance experiments and I am not showing it in this figure, but we have some small cylinders of silicon that we use for pulse-echo measurements, so there were 3 kinds of measurements made.

DR. SMITH: How big are those?

DR. LEDBETTER: Our cylinders were about 1.5 cm in diameter and a centimeter thick.

[Transparency 4]

This shows the fixture arrangement for measuring the spheres. It is what I call a tripod with 3 transducers. The only force is the gravitational force.

[Transparency 5]

Here is the usual, almost perfunctory, spectrum. I just want to emphasize, since we are going to talk about internal friction, that there are various ways to get it. We get it the standard way from the half-power width, $\Delta f/f$.

[Transparency 6]

Just as we try to get the complete real part of the elastic stiffness, we try to get the complete imaginary part, the internal friction. I do not want you to read these numbers (you probably cannot), but for these 5 spheres these are the measurement results.

You can go down the left-hand column and pick out your favorite elastic constant and go across and see how it changes. The numbers at the bottom are the Kröner model quasi-isotropic averages for the bulk modulus, shear modulus, and so on.

I forgot to bring a viewgraph that shows an average of these results compared with the old McSkimin (Bell Laboratories) results from the 1960s. They are the same with one part in 1000.

[Transparency 7]

If we turn to the internal friction and if we measure it versus strain, we get what was for us a rather startling curve. Right away this tells you a lot of things. One is that while silicon may be a good material for an international mass standard and probably a very good material for velocity for an elastic-stiffness standard, it is a very poor material for an internal friction standard, because, depending on your measurement conditions, you get different results.

PARTICIPANT: Is that the strain amplitude of your vibration? What is the strain?

DR. LEDBETTER: That is the strain in the crystal induced by the piezoelectric transducer.

We plotted internal friction versus the strain and it was just completely unexpected, because we have a variation of a factor of 3 or 4 between the lowest and highest values and we have, obviously, 1, 2, 3, 4 phases or processes contributing to internal friction.

We wondered how can we explain this? As Dr. Ogi mentioned, oftentimes for most materials the largest contributions to internal friction are dislocations, so we looked at that first.

[Transparency 8]

It is useful to look at a simple equation that comes out of the Koehler-Granato-Lucke vibrating string theory for vibrating dislocations. Most of these are basically material parameters. Important parameters for our purposes are gamma, the dislocation density, and L, the dislocation length. Those are things I shall be talking about, and how dislocation properties affect Q inverse.

This is a low-frequency equation where the Q inverse goes as frequency.

[Transparency 9]

If we make a schematic of the Granato-Lucke model for higher frequencies, we find that the internal friction goes through a maximum and then it has a 1/omega dependence, but the important thing is that the frequency of this maximum goes as $1/B\ell^2$. This ℓ is the same as the L in the previous viewgraph; it is the dislocation length. B is the viscosity seen by the dislocations as they vibrate. Almost all the materials that we have measured show this kind of dependence on frequency, a linear increase.

[Transparency 10]

We measured these materials and we found the following results: Cumulative internal friction versus frequency decreased. Here is shown the Granato-Lücke slope, which approximately fits our results.

I should have said, when I showed the previous graph with the big wide resonance, that for a typical material, let's say copper, that maximum occurs at about 1 to 10 MHz and because it depends on $1/L^2$, to increase L by a factor of 10 and drops that frequency considerably.

What these results suggest is that we are on the high side of the curve and the maxima is somewhere below 400 kHz, simply because in these materials we have few dislocations, and we have few impurity atoms that serve as pinning points for dislocations.

[Transparency 11]

This is taken from the very old literature, some really almost visionary work by Thomas Read. In 1940, when most people in the world did not believe in dislocations, he was studying them. He discovered this strain effect. If we measure internal friction versus strain, we get 2 components, a strain-independent part and a strain-dependent part.

This strain-dependent part, of course, results from depinning the dislocations and increasing the loop length. This, remember, contributes to low frequencies as L^4. This is very helpful but obviously does not explain these four regions we have in our measurement results on the silicon.

[Transparency 12]

We wondered what about kinks, kinks and dislocations. This has been a big controversy since the 1950s -- I think the idea was conceived by a professor who worked on it at Northwestern but then taken up vigorously by Seeger at Stuttgart and this argument rages to this day and we have these two camps, almost with no dialogue.

For those of you who do not know what a kink is, we have a dislocation lying in a Peierls valley and if we apply a stress on it to move it, it cannot move, because the stress is less than the potential associated with the Peierls hill, but one way to make it move is to somehow get a small part of the dislocation over and then move this part, this kink, either left or right, depending on which way the force is, so it is much easier, then, and, by moving this, let's say, in this direction, then this entire dislocation moves from one valley to the next.

So then we had to wonder, well, if there are kinks, how would they affect the internal friction?

[Transparency 13]

Fortunately, actually, when I was in graduate school, I met Tetsuro Suzuki, who was a postdoc from Japan and I knew about this paper he had published with Elbaum when Suzuki was at Brown University, and they actually considered this question: If we have kinks, what is the strain dependence of the internal friction?

That is what they plotted here. They say at very low strains -- I am not sure you can read this (the lowest strains shown is 10^{-10}, and it goes 10^{-8}, 10^{-6}) -- there is a plateau. At some strain, which they calculate to be around 5×10^{-5}, Q^{-1} goes to 0. It goes to 0 because it is sort of an

exhaustion process. As these kinks move out to the ends of the dislocations -- well, they are effectively just frozen out of the deformation process.

So this was very useful, because then we could arrange them in our work, our measurements, and say the following: For low strains we have this kink plateau, we have this kink internal friction going toward 0, and about where it does we have the Granato-Lucke and the depinning process starting, and usually this curve just goes up rather indefinitely. So why does it bend over?

[Transparency 14]

It is because of this special material. There are very few pinning points. There are very long loop lengths. You really, I think, literally run out of pinning points; after a certain point there are no more dislocation pin points to break, so that is what is happening here, we are exhausting these.

We have not done any quantitative modeling of this and I think it is possible, and I also think it is possible to at least establish bounds on some of these dislocation parameters, like the dislocation density, like the dislocation length, and maybe dislocation viscosity, which still is quite uncertain despite much good research.

[Transparency 15]

Here is a viewgraph almost everyone likes to see, one with conclusions. I wrote down three. First of all, within about one part in one thousand (I did not show you all the old measurements) what I have called the new silicon elastic stiffness measurements agree with the old ones. It seems at first quite trivial but it is not trivial for two reasons.

One, there are people who really are working with these silicon devices, they really at least think they need to know the best possible numbers and, secondly, in some materials, for example, in monocrystal copper that we have measured, we can measure one purity of pure copper and one with parts per million doping and get a 2% difference in some of the elastic stiffnesses, so some materials have a very large difference.

As I said, at low strains we think the Q inverse is dominated by kink movement, which means to see this you must measure at low strains. At high strains, the effect discovered by Read and published in 1940 and then elaborated by Granato and Lucke in *The Journal of Applied Physics* in about 1955 comes in and we see that mechanism that then saturates. Thank you.

DR. SACHSE: I am not as optimistic as you that you will ever be able to do quantitative comparison. There are just too many variables. The free parameter you mentioned, that is really not well known, the loop length is not well known, the density is not well known, so that is one part of the picture; there are just so many parameters.

The second thing is, you plotted the Q - 1 versus the strain and I am not sure exactly what you mean by that. Someone else asked, "Is that the driving amplitude?"

DR. LEDBETTER: No, it was the strain in the specimen.

DR. SACHSE: Now, if this is a resonating specimen, that means the strains are not uniform in this specimen. You are going to have to worry, then, about the nonuniformity of the strain and how that affects the dislocations. It sounds like a can of worms to me to try to get a quantitative measure of what is going on there, because there will be strains on some parts of the sample that are not vibrating at large amplitudes at all, so how will you do the integration? It is interesting but I just do not know how to cope with it.

DR. LEDBETTER: I do not know how familiar you are with the literature. There has been an enormous amount of work and people over the years have generated these big broad resonance curves I showed you, you get low frequencies and high frequencies and occasionally right at the top, and some people have gone to the trouble to count and check the dislocations and this is a very difficult process.

All I can say is the model, based on the literature, has enormous acceptance and wide application, but how many variables are there? There is the density and the length. I think I did do some calculations on this and the numbers were not unreasonable, they are not imaginary, not negative, and not off by 2 orders of magnitude.

I think I share your unease but I think when we sit down and look at this, it is a little more positive.

MIGLIORI: Remind me again how you measured the strain amplitudes.

DR. LEDBETTER: You should see page 82 of Migliori and Sarrao. (Laughter)

DR. MIGLIORI: I was uneasy when I wrote that. (Laughter)

DR. LEDBETTER: You raise an interesting question and I think there really remains some question about what is the absolute number of the strain. Last year we tried to get someone going with a dynamic interferometer device to measure strains, but it did not go well, the program, and I was not sure what I would do with the results anyway, so I sort of let it go.

DR. MIGLIORI: So you use the Q and the known displacement of the transducer, roughly.

DR. LEDBETTER: And the internal friction of the specimen.

PARTICIPANT: You mentioned dislocation and length as one of the parameters. Does that imply that there is a fairly narrow distribution of planes or is that distribution also --

DR. LEDBETTER: The early model considered just one length. The later models brought variable length into a spectrum. For present purposes I assumed just this one length.

PARTICIPANT: Maybe related to it, I am not sure, it seems that this depinning occurs over a fairly narrow range of strains, that everything is over with in just a fairly narrow range. Is that significant, perhaps? I guess I wonder if you could comment on that.

DR. LEDBETTER: I had not thought about it, but it does not surprise me. After all, we are talking about trying to move dislocations or parts of dislocations through a lattice against some barrier and at a certain stress level they will not move and at higher stress they will, kind of a yield point in a sense, at a micro-level.

DR. MARSTON: To go back to the question of the strain field because you are driving a particular mode, is it understood in either the kink model or the Lücke model how the dislocations behave, how the position dependences -- in other words, suppose you are at a strain anti-node versus a node, is it understood how that dislocation varies with where the dislocations are?

DR. LEDBETTER: I am not sure.

DR. MARSTON: That would seem as if, if you had an understanding of that, that you could answer his question as opposed to some other ways these models may be tested with traveling waves where that issue would not come up.

DR. LEDBETTER: Yes, I am not sure.

DR. SACHSE: May I make a comment? You have to remember what the Granato-Lucke model is. Essentially they treat the dislocation as a string pinned at the ends and then they have that B value, which is the drag. That is the model. It is a very simple model and then they analyze this and crank out from that what is the end.

DR. LEDBETTER: It is interesting, the equation of motion for this is so widespread through solid-state physics. You can probably find 30 phenomena in some list of things that follow this same equation of motion; it is just an equation for a damped harmonic oscillator, basically.

DR. HICKEY: Since we have some extra time, Dr. Ledbetter wants to present some new results of some measurements, I believe.

DR. LEDBETTER: Yes, these really are new results.

[Transparency 16]

We got these about one week ago. We thought they were quite interesting. It is really our first start in considering piezoelectric crystals. The first author is Ming Lei, who is at DRS Company in Wyoming, the same as Dr. Willis. His contribution was that he did most of the analysis and repeated our measurements that we made in Boulder.

[Transparency 17]

Langasite may not be a material that you have heard about. It has been known for some years, especially in Russia; if you do a literature search, probably 9 out of 10 papers will be Russian. Especially they see it as a replacement material for quartz, which is really astonishing, because, according to good sources, each year there are 2 billion quartz devices made.

So what would be langasite's possible advantages? One, it does have higher piezoelectric coefficients. Secondly, it has no phase transformation. Quartz has a phase transformation at 573°C. Thirdly, there is sort of a hope, it has lower dielectric loss. As far as I know this has not been resolved yet.

[Transparency 18]

In this table I want to show you basically two things. I want to give you the chemical composition of langasite. It is $La_3Ga_5Si0_{14}$. It is interesting in terms of its crystal symmetry. It is trigonal, and has the same point group and its space group as quartz.

It is rather different from quartz, as you might expect from the chemical formulas, in that it has 23 atoms per unit cell, whereas quartz has 9.

People are now growing large numbers of single crystals.

[Transparency 19]

One of these crystals is shown in the measurement fixture. It was a cylinder about 1 cm in diameter and 2-3 cm long. As a government employee, I am not supposed to tell you what commercial apparatus we use, but anyway -- (Laughter) -- when Ming Lei measured these, he measured them three ways, edge/edge, end/end. Then, he set up a horizontal 4-point probe. All methods gave essentially the same results, he told me.

[Transparency 20]

Here is the spectrum for quartz. Just as before, we get the internal friction from this half-width.

DR. MIGLIORI: You did not give the elastic constants for the trigonal structure? On the previous viewgraph did you determine the elastic constants as well as the Q? You measured frequencies and Q's. Did you use the frequencies to fit elastic moduli for the trigonal single crystal?

DR. LEDBETTER: Yes, I am going to talk about the elastic and piezoelectric models.

[Transparency 21]

For the quartz case, if you look in the literature, there are lots and lots of measurements, 8 or 10 good measurements. What I am showing here is our results compared with Ohno's results using a parallelepiped -- again, the old Bell Laboratory results, and they are essentially the same.

It is especially important that they are the same for C_{14}, an elastic constant usually zero. It is nonzero in trigonal crystals and is very troublesome, both for people making measurements and for people doing any analysis or theory. Especially if you look in the literature, these numbers just vary enormously. For these three cases, at least, there seems to be some consensus about it.

[Transparency 22]

Here is the quartz compared to langasite. We see, first of all, langasite is a much more dense material. As we might expect, these longitudinal moduli, C_{11} and C_{33}, are different by approximately the density ratio, but it is interesting (I do not quite understand why yet) that the shear moduli are roughly the same.

[Transparency 23]

Here I want to distinguish between the elastic material and the piezoelectric material and there are many ways to do this. This is just the Christoffel equation, so the eigenvalues are rho v^2 and these eigenvalues depend, for the elastic model, only on the elastic-stiffness coefficients, but in the piezoelectric case they depend additionally on the piezoelectric coefficients, epsilon$_{rij}$ and the dielectric coefficients K_{mn}.

In principle, if you are making an RUS measurement, you should take these into account, but it also offers a possibility of using RUS measurements to determine these electromechanical quantities in addition to these elastic (this was one of our main initial interests).

[Transparency 24]

However, for the quartz case, we plot the predicted frequencies from the determined elastic stiffnesses versus the measured frequencies, and we get essentially a perfect line for the elastic model. In other words, this assumes no piezoelectricity. What it means, conversely, is, unless you have a very sophisticated measurement system (better than ours), you probably cannot get any piezoelectric coefficients from these kinds of measurements.

I think in Professor Ohno's paper on quartz he reached the same conclusion.

[Transparency 25]

The langasite results look essentially the same as the quartz. The elastic model has a perfect linear dependence; we cannot extract information about the piezoelectric coefficients.

[Transparency 26]

However, this is not true for all materials. Here are some results for lithium niobate, also a trigonal material. The open circles represent the elastic analysis, which is not on this 45° line, but when we bring in the known measured, piezoelectric coefficients and do the calculation, we get quite good agreement.

For this material, it seems, or things with similar dielectric coefficients, one could use RUS, in principle, not only to measure the elastic stiffnesses but also the piezoelectric coefficients.

[Transparency 27]

There are not many conclusions about this, since these results are about two weeks old, but here are our current plans. We are interested in focusing on the internal friction of things like quartz and perhaps langasite, especially relating it to the defect mechanism. What is the defect mechanism? I know this is an old subject with papers by Mason of 30 years ago but, nevertheless, I am not sure it is settled.

Secondly, we are quite interested in the relationship between the mechanical Q and the electrical Q. I mean, if the dipole defect is both an elastic dipole and an electric dipole, then one can probably make some simple model connecting these two terms, and we think this would be quite useful, because I think RUS measurements are much easier to make than most of the dielectric loss measurements that I see made at our laboratory.

As I just mentioned a few minutes ago, these materials that have enough coupling in the piezoelectric strain we can actually see if we can derive piezoelectric coefficients from an RUS spectrum.

DR. MIGLIORI: The distinction between the electronic and the mechanical Q, let's see, you would have to turn off the electric fields in order to measure the mechanical Q.

DR. LEDBETTER: Those would be 2 different experiments in 2 different parts of the world. I would not care.

DR. MIGLIORI: Yes, but the point being that when this stuff is vibrating mechanically it has an electric field or electric currents that are driven by the mechanical vibrations, so it is hard to see how you separate it in 2 pieces.

DR. LEDBETTER: I do not understand. I mean, we measure mechanical loss, some fellow in southern China measures electrical loss, and we make some model connecting the two.

DR. MIGLIORI: But the Q ought to be the same in either case and it is not.

DR. LEDBETTER: No, why should it be the same? It is coefficients, remember? I can guarantee you they are not the same.

DR. MIGLIORI: But the full width at half-maximum for the resonances -- you are on mechanical resonance when you are driving them electrically using it as a quartz crystal oscillator, so it is different.

DR. LEDBETTER: I do not think so. I do not think the response to an elastic dipole is the same as the response to an electric dipole. I think they are proportional in many cases, probably.

DR. MIGLIORI: But not the same?

DR. LEDBETTER: Not the same, no. [Added after workshop: Imagine an elastic dipole without charge, which gives a certain Q^{-1} (mech), but Q^{-1} (elec) = 0. Then, we add charge, Q^{-1} (mech) is unaffected, but Q^{-1} (elec) becomes nonzero.]

Thank you.